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⑭発明の名称　燃料電池

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㉑発明者　岡　　篤　弘　神奈川県川崎市川崎区田辺新田1番1号　富士電機株式会社内
㉑出願人　富士電機株式会社　神奈川県川崎市川崎区田辺新田1番1号
㉔代理人　弁理士　山口　巌

明　細　書

1.発明の名称　燃料電池

2.特許請求の範囲

1）電解質としてのりん酸を保持するマトリックスと、このマトリックスの両面に密着して配された燃料電極および酸化剤電極とを有する単位セル構成層を単位セル相互間にガス不透過性のセパレータを介在させて積層したものからなり、前記一対の電極が前記セパレータ側に反応ガスの通路となる溝を有するガス不透過性のリブ付電極基材と、このリブ付電極基材の前記マトリックス側に担持した電極触媒層とからなるものにおいて、前記溝に沿って前記反応ガスの入口側でガス透過性が低く、反応ガスの出口側でガス透過性が高いガス透過性の制御手段を有するリブ付電極基材を備えてなることを特徴とする燃料電池。

2）ガス透過性の制御手段が、溝の底部におけるリブ付電極基材の厚みが反応ガスの出口側に向けて小さくなるよう前記溝の底面を反応ガスの出口側に向けて傾斜して形成したものであることを特徴とする請求項1記載の燃料電池。

3）ガス透過性の制御手段が、リブ付き電極基材のカーボン繊維の密度を溝に沿って反応ガスの入口側で密に、その出口側で疎になるよう形成したものであることを特徴とする請求項1記載の燃料電池。

3.発明の詳細な説明

〔産業上の利用分野〕

この発明は、リブ付電極基材方式の燃料電極および酸化剤電極を有する積層形の燃料電池、ことに反応ガス通路に沿った方向の発電特性の分布を均等化したリブ付電極基材の構造に関する。

〔従来の技術〕

第6図はリブ付電極方式による従来の燃料電池の一つの単位セル部分を示す斜視図である。図において、単位セル1は電解質としてのりん酸を保持するマトリックス2と、マトリックス2を挟んでその両側に配された燃料電極3および酸化剤電極4の積層体として構成される。各電極3および4はそれぞれ一方の面が角溝状に形成されたガス

透過性を有するリブ付電極基材５と、その平滑な面側に形成された電極触媒層６とで成層されており、かかる単位セル１対以上を順次にガス不透過性のセパレータ７を介在させて積層することによりセルスタックが構成される。

また、電極基材５に角度によって形成される溝５Ａ，５Ｂはその溝が互いに直交するよう配置され、セパレータ７との間に溝数の互いに並列した燃料ガス通路５Ａおよび酸化剤ガス通路５Ｂが形成される。また、セルスタックの４つの側壁面には図示しないマニホールドが気密に取りつけられ、マニホールドを介して燃料ガス通路５Ａには水素リッチな燃料ガス８Ｆが、また酸化剤としての反応空気８Ａが供給される。

供給された燃料ガス８Ｆ，反応空気８Ａ等の反応ガス８は、反応活動質である水素または酸素がガス透過性の電極基材５中を拡散によって透過してマトリックス２との界面に到達し、りん酸で満たされた電極触媒微粒子と接触して三相界面を形成し、ここで電気化学的反応に基づく直接発電が行われる。

［発明が解決しようとする課題］

リブ付電極基材５の溝５にその一方端から導入し、他方端から排出される燃料ガス，反応空気等の反応ガス８は、溝内を進行する過程で反応の活動質である水素または酸素が電極基材中に拡散するのでその濃度が徐々に低下する。このとき、活動質が基材中に拡散する速度は反応ガス中の活動質濃度に比例するので、基材を透過して三相界面に到達する活動質の分圧は第５図に曲線１０１で示すように、反応ガスの入口側で高く，出口側で低い不均等分布を示す。その結果、反応ガスの入口付近では拡散速度が高くしたがって濃度過電圧に基づく電圧降下が小さいため、高い動作電圧を保持するが、出口付近では拡散速度が低下して濃度過電圧が増すために動作電圧が低くなり、電極基材５の溝に沿って電位差が発生する。また、電極基材５には電位差を緩和する方向（溝５Ａ，５Ｂに沿った面方向）に電流が流れ、これによって抵抗過電圧が増加する。このように従来の燃料電池では反応ガス通路に沿って動作電圧に不均等分布が生じ、さらにこれが原因で電極面積を有効に活用できないために発電性能が低下するなどの問題点が発生する。

この発明の目的は、リブ付電極基材の構造を改善することにより、三相界面における活動質の濃度分布を均等化することにある。

［課題を解決するための手段］

上記課題を解決するために、この発明によれば、電解質としてのりん酸を保持するマトリックスと、このマトリックスの両面に密着して設けられた燃料電極および酸化剤電極とを有する単位セル複数層を単位セル相互間にガス不透過性のセパレータを介在させて積層したものからなり、前記一対の電極が前記セパレータ側に反応ガスの通路となる溝を有するガス透過性のリブ付電極基材と、このリブ付電極基材の前記マトリックス側に担持した電極触媒とからなるものにおいて、前記溝に沿って前記反応ガスの入口側でガス透過性が低く，反応ガスの出口側でガス透過性が高いガス透過性の制御手段を有するリブ付電極基材を備えてなるものとし、具体的にはガス透過性の制御手段が、溝の底部におけるリブ付電極基材の厚みが反応ガスの出口側に向けて薄くなるよう前記溝の底面を反応ガスの出口側に向けて傾斜して形成したもの、またはガス透過性の制御手段が、リブ付き電極基材のカーボン繊維の密度を溝に沿って反応ガスの入口側で密に，その出口側で疎になるよう形成したものとする。

［作用］

この発明において、溝に沿って反応ガスの入口側でガス透過性が低く，出口側でガス透過性が高いガス透過性の制御手段を有するリブ付電極基材を備えるよう構成したことにより、溝の出口側に向けて低下する反応ガス中の水素，酸素等の活動質の濃度分布を、これとは逆に出口側に向けて高くなる電極基材のガス透過性によって補償することが可能となり、電極面積を有効活用する効果と、燃料電池の動作電圧の不均等分布を防止する効果とを同時に得ることができる。また、ガス透過性

の制御手段としては、溝をその出口側に向けて深くして溝の底部の電極基材の厚みを出口側に向けて薄くする方法、あるいは電極基材のカーボン繊維の密度を溝の出口側に向けて疎くしてガス透過性を制御する方法のいずれによっても目的す機能を得ることができる。

〔実施例〕

以下この発明を実施例に基づいて説明する。

第1図にこの発明の実施例になる燃料電池におけるガス透過性の制御手段を示す要部の断面図、第2図および第3図は第1図におけるA－A位置およびB－B位置それぞれの要部の断面図である。図において、電解質膜層6とセパレータ7との間に位置するリブ付電極基材15には、その溝25の深さdが反応ガス8の入口側で小さく（d_1）、出口側で大きく（d_2）なるよう溝の底面25Bが溝に沿って傾斜するよう形成される。これによって溝の底面25Bと電極触媒層6との間の電極基材の厚みtは入口側でt_1が厚く、出口側でt_2が薄く形成されることになり、この部分での反応活物質としての水素または酸素の透過性が反応ガス8の入口側で低く、出口側で高い分布を持ったガス透過性の制御手段が形成される。

前述のように実施例によれば、溝の入口側に流入した反応ガス8は活物質の濃度は高いものの、厚みtなる基材の厚みが厚くガス透過性が低いために、これを拡散によって透過して三相界面に達する活物質の分圧が抑制される。また、溝の出口側近くに到達した反応ガス中の反応活物質の濃度は途中で活物質が消費されることによって入口側より低くなるが、厚みtが薄い基材のガス透過性が高いために、基材を透過して三相界面に達する反応活物質の分圧の低下は入口側よりも少くなる。その結果、三相界面における反応活物質としての水素または酸素の分圧は第5図に曲線110で示すように、従来の分布曲線101に比べて傾斜がゆるくなり、したがって溝に沿った方向の発電反応の分布が従来よりも均等化されるので、電極面積を発電反応に有効に利用して分極による動作電圧の低下の少い燃料電池を得ることができる。

第4図はこの発明の異なる実施例におけるガス透過性の制御手段を示す要部の断面図であり、リブ付電極基材35はその溝5′または5Aの深さdが従来のそれと同様に一様に形成され、したがって溝の底部における基材の厚みtも溝に沿って一様な厚みを保持するが、この部分におけるカーボン繊維の密度が反応ガス8の入口側で密に、出口側で疎になるよう、密度に変化を持たせることによってガス透過性の制御手段を構成した点が前述の実施例と異なっている。このように構成したリブ付き電極基材35の溝に沿った方向のガス透過性は、反応ガスの入口側で低く、出口側に向かって高くなる分布特性を持つので、電極基材を透過して三相界面に達する水素、酸素等の反応活物質の分圧を前述の実施例と同様に、溝に沿って均等化し、したがって発電反応の分布を改善し、電極面積を有効活用できるとともに、動作電圧の不均等分布も改善することができる。

〔発明の効果〕

この発明は前述のように、反応ガスの流通路となる溝を反応ガスの出口側に向けて深くするか、あるいはカーボン繊維の密度を反応ガスの出口側に向けて疎にするなどの手段により、反応活物質としての水素または酸素の透過性が反応ガスの入口側で低く出口側で高いガス透過性の制御手段を有するリブ付電極基材を備えるよう構成した。その結果、反応ガス通路に流入した反応ガス中の活物質の濃度が通路の出口側に向けて低くなることによって三相界面に生ずる活物質の分圧の不均等分布が、これと逆の分布特性を持つガス透過性の制御手段によって補償され、三相界面における活物質の分圧分布を均等化できるので、分圧の不均等分布が原因で従来問題となった三相界面での発電反応の不均等分布、およびこれに起因して燃料電池に生ずる動作電圧、電流の不均等分布の問題点や、電極面積が有効に活用されないために生ずる発電性能の低下などの問題点が解決され、したがって電極面積全体を有効に利用して優れた発電性能を安定して得られる燃料電池を提供することが

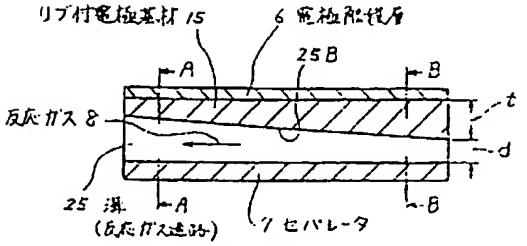
できる。

4.図面の簡単な説明

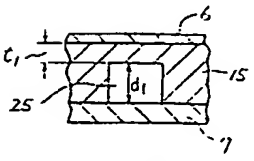
第１図はこの発明の実施例になる燃料電池のガス透過性手段を示す断面図、第２図は第１図におけるＡ－Ａ位置の断面図、第３図は第１図におけるＢ－Ｂ位置の断面図、第４図はこの発明の他の実施例を示す断面図、第５図は実施例における反応物質の分圧分布を従来技術のそれと比較して示す特性図、第６図は単位セルの一般的構成を示す斜視図である。

１…単位セル、２…マトリックス、３…燃料電極、４…酸化剤電極、５，１５，３５…リブ付電極基板、６…電極触媒層、７…セパレータ、８Ｆ，８Ａ，８…反応ガス、５Ａ，５Ｂ，２５…溝（反応ガス通路）、２５Ｂ…溝の傾斜した底面、ｄ…溝の深さ、ｔ…電極部の電極基材の厚み。

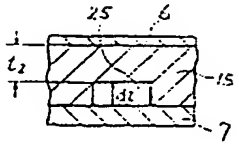
代理人弁理士　山口　巌



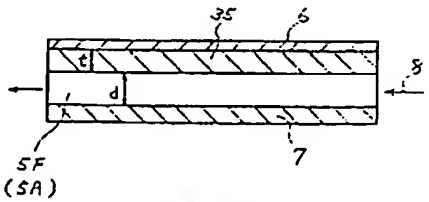
第１図



第２図



第３図



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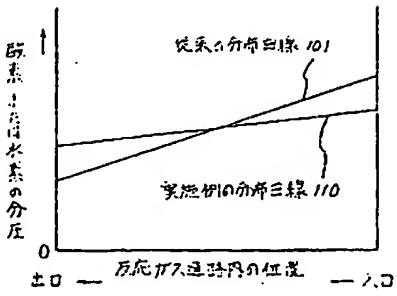
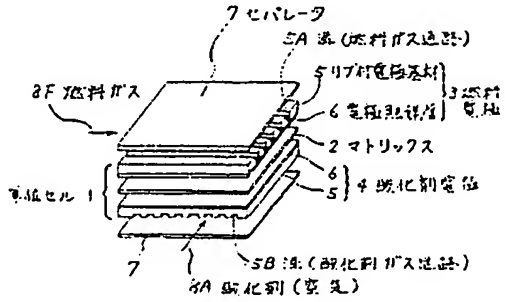


第５図



第６図

======= PAJ =======

TI - FUEL CELL

AB - PURPOSE:To unify the concentration distribution of an active material on the three-phase interface by providing a ribbed electrode substrate having a control means of the gas permeability which is low on the inlet side of the reaction gas along a groove and high on the outlet side.
- CONSTITUTION:The bottom face 25B of a groove 25 is slantly formed along the groove 25 on a rib electrode substrate 15 located between an electrode catalyst layer 6 and a separator 7 so that the depth (d) of the groove 25 is made small (d2) on the inlet side t2 of the reaction gas 8 and large (d1) on the outlet side t1. The thickness (t) of the electrode substrate 15 between the bottom face 25B of the groove 25 and the electrode catalyst layer 6 is made thick on the inlet side t2 and thin on the outlet side t1, thus a control means of the gas permeability of hydrogen or oxygen as a reaction active material at this portion which is low on the inlet t2 side of the reaction gas 8 and high on the outlet t1 side is formed. The concentration distribution of the active material generated on the three-phase interface is unified.

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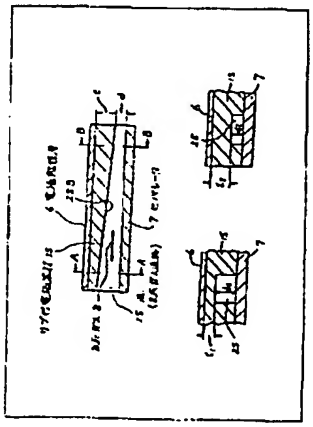
ABD - 19920309
ABV - 016095
AP - JP19900076211 19900326
GR - E1175

PA - FUJI ELECTRIC CO LTD
IN - OKA YOSHIHIRO

I - H01M8/02



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(21) Application Number: H2-76211
(22) Date of Application: 26 Mar 1990
(72) Inventor: Yoshihiro OKA of Fuji Electric Co. Ltd. 1-1, Tanabe Shinden, Kawasaki-ku, Kawasaki-shi
(71) Applicant: Fuji Electric Co. Ltd. 1-1, Tanabe Shinden, Kawasaki-ku, Kawasaki-shi
(74) Patent attorney: Iwao YAMAGUCHI

1) Title: Fuel Cell

2) Claims:
1. A fuel cell including a plurality of single cells, each cell having a matrix holding a phosphoric acid electrolyte, and a fuel electrode and an oxidising agent electrode disposed in a close-fitting manner on each side of said matrix, whereby a plurality of single cells form a stack with gas impermeable separators interposed between each cell, and said electrodes comprise a gas impermeable ribbed electrode substrate having channels forming a reaction gas passage on one side of said separator, and an electrode catalyst layer supported on the matrix side of the ribbed electrode substrate, wherein the ribbed electrode substrate is provided with means for controlling the gas permeability such that the gas permeability along said channels is low on the reaction gas inlet side, and high on the reaction gas outlet side.

2. A fuel cell as recited in Claim 1,
wherein said means for controlling the gas permeability is provided by reducing the thickness of the ribbed electrode in the bottom part of the channels in the direction of the reaction gas outlet side so that the bottom surface of said channels is inclined towards the reaction gas outlet side.

3. A fuel cell as recited in Claim 1,
wherein said means for controlling the gas permeability is provided by a structure in which the density of carbon fibres in the ribbed electrode substrate is high along the channel at the gas inlet side, and reduces towards the outlet side.

3) Detailed description of the Invention

Industrial Application

This invention relates to stacked-type fuel cells having a fuel electrode and an oxidising agent electrode in a ribbed electrode substrate system, and relates especially to a ribbed electrode structure which evens out the power generation characteristic along the reaction gas passage.

Prior Art

Fig. 6 is a perspective view of a single cell of a conventional fuel cell using a ribbed electrode system. In the drawing, the single cell 1 comprises a matrix 2 supporting a

phosphoric acid electrolyte, the matrix 2 being interposed between a fuel electrode 3 and an oxidising agent electrode on respective sides to form a layered body. The electrodes 3 and 4 are formed from a gas-permeable ribbed electrode substrate 5 having a corrugated shape on one side and an electrode catalyst layer 6 formed on the smooth side. Gas impermeable separators 7 are interposed between single cells 1 to form a multilayer stack.

The arrangement of the respective channels 5A,5B formed in the electrode substrate 5 by the corrugations is mutually orthogonal, and the a plurality of mutually parallel fuel gas passages 5A and oxidising gas passages 5B are formed between the separators 7. A manifold (not shown) is gas-tightly attached to the four side walls of the cell stack. Hydrogen rich fuel gas 8F and a reaction air 8A acting as an oxidising agent are supplied to the fuel gas passage 5A via the manifold.

The active substances hydrogen and oxygen in the supplied fuel gas 8F and the reaction gas 8 such as the reaction air 8A permeate the gas permeable electrode material by diffusion and reach an interface with the matrix 2, and make contact with electrode catalytic particles that are impregnated with phosphoric acid, to form a three-phase interface. Electric power is directly generated as a result of the electrochemical reaction.

Problem to be solved by the invention

The fuel gas and the reaction gas 8 such as reaction air flow into the channels from one end of the ribbed electrode substrate 5, and are exhausted from the other end. During the progression along the channels, the active substances hydrogen and oxygen diffuse into the electrode substrate, so that their concentration gradually decreases. At this time, the rate at which the active substances diffuse into the substrate is proportional to the concentration of active substances in the reaction gas. The partial pressure of the active substances that reach the three-phase interface after penetrating the substrate is shown by curve 101 in fig. 5, and the distribution is uneven, the partial pressure being high at the reaction gas inlet side and low at the outlet side. Consequently, near the reaction gas inlet, the diffusion rate is high, the voltage drop is small due to the concentration overvoltage, and a high operating voltage is maintained. However, near the reaction gas outlet, the diffusion rate reduces, the concentration overvoltage increases and the operating voltage reduces, creating a potential difference along the channels of the electrode substrate 5. In the electrode substrate 5, current flows in a direction that will lessen the potential (edgewise along channels 5A, 5B), and causes the resistive overvoltage to increase. Therefore, in a conventional fuel cell, the operating voltage has an uneven distribution along the reaction gas passage. Because of this, the electrode area cannot be efficiently used. and the power generating performance falls.

An aim of the present invention is to improve the structure of the ribbed electrode substrate so that the concentration distribution of active substances at the three-phase interface is made uniform.

Means for solving the problem

To solve the above-mentioned problem, a fuel cell is provided comprising a plurality of single cells, each cell having a matrix holding a phosphoric acid electrolyte, and a fuel electrode and an oxidising agent electrode disposed in a close-fitting manner on each side of said matrix, whereby a plurality of single cells form a stack with gas impermeable separators interposed between each cell, and the pair of electrodes comprises a gas impermeable ribbed electrode substrate having channels forming a reaction gas passage on one side of the separator, and an electrode catalyst layer supported on the matrix side of the ribbed electrode substrate, wherein the ribbed electrode substrate is provided with means for controlling the gas permeability such that the gas permeability along the channels is low on the reaction gas inlet side, and high on the reaction gas outlet side. Specifically, the means for controlling the gas permeability is provided by inclining the bottom surface of the channels towards the reaction gas outlet side by reducing the thickness of the ribbed electrode in the bottom part of the channels in the direction of the reaction gas outlet side. Additionally, the means for controlling the gas permeability is provided by packing carbon fibres of the ribbed electrode substrate tightly along the channel at the gas inlet side, and more loosely towards the outlet side.

Operation

In accordance with the present invention, a ribbed electrode substrate is provided with means of controlling the gas permeability such that the gas permeability along the channels is low at the reaction gas inlet side and high at the outlet side. The reduction in the concentration distribution of the active substances hydrogen and oxygen in the reaction gas towards the outlet side is thus compensated by the increasing gas permeability of the electrode substrate towards the outlet side. In addition to the efficient use of the electrode area, an uneven distribution of the operating voltage of the fuel cell can also be prevented. Further, as a means for controlling the gas permeability, a method can be employed in which the thickness of the bottom part of the channels in the electrode substrate is reduced towards the outlet side, or a method which controls the gas permeability by reducing the density of carbon fibres of the electrode substrate towards the outlet side of the channels.

Embodiments

An embodiment of the present invention is described below.
Fig. 1 is a sectional view of the main part of a gas permeability control means in a fuel cell in accordance with an embodiment of the present invention.
Figs. 2 and 3 are sectional views of the main part shown in fig. 1 along A-A and B-B respectively.
With reference to the drawings, ribbed electrode 15 is positioned between electrode catalyst layer 6 and separator 7, and provided with channels 25 whose depth reduces to d_2 at the reaction gas 8 inlet side, and increases to d_1 at the inlet side, so that the bottom surface 25B of the channels is inclined along the channels. Accordingly, the thickness t of the electrode substrate between the bottom surface 25B of the channels and the electrode catalyst layer 6 is large (t_2) at the inlet side and small (t_1) at the outlet side, thus providing a means of controlling the gas permeability with a distribution that has a low permeability to the active substances hydrogen and oxygen at the inlet side, and a high permeability at the outlet side.

In accordance with the embodiment described above, the reaction gas 8 flowing into the channels at the inlet side has a high concentration of active substances, but since the substrate thickness is large (t_2), the gas permeability is low, and the partial pressure of the active substances reaching the three-phase layer by diffusing through the substrate is suppressed. Although the concentration of active substances in the reaction gas approaching the outlet side of the channels has reduced en route, and is less than it was at the inlet side, because the gas permeability of the substrate that has a low thickness (t_1) is high, the reduction in partial pressure of the active substances penetrating the substrate and reaching the three-phase interface is less than at the inlet side. As a result, the curve 110 (fig. 5) of the partial pressure distribution of the active substances hydrogen and oxygen at the three-phase interface is has a more gentle slope compared with the prior art distribution curve 101. Consequently, the distribution of the electric power generating reaction in the direction of the channels is more uniform than in the prior art, so that the electrode area can be used more efficiently for the power generation reaction, and a fuel cell can be provided in which the reduction in operating voltage due to polarisation is low.

Fig. 4 is a sectional view of the main part of a gas permeability control means in accordance with a further embodiment of the present invention. Ribbed electrode 35 is provided with channels 5F or 5A whose depth d is constant, as in the prior art, and the thickness t of the substrate at the bottom part of the channels is maintained along the channels. However, the density of carbon fibres varies so that it is high at the reaction gas inlet side and low at the outlet side, forming a gas permeability control means having a different structure from that of the previous embodiment. The gas permeability along the channel direction of the ribbed electrode substrate 35 of this structure has a distribution characteristic in which the permeability is low at the reaction gas inlet side and increases towards the outlet side. As in the previous embodiment, the partial pressure of the active substances hydrogen and oxygen reaching the three-phase interface after penetrating the electrode substrate is therefore evened out along the channels. Consequently, the distribution of the electric power generating reaction is improved, the electrode area is used efficiently, and the uneven distribution of the operating voltage is improved.

Effect of the invention

As described above the present invention provides a ribbed electrode substrate in which the channels forming the reaction gas fluid passage become deeper towards the reaction gas outlet side, or in which the density of carbon fibres reduces towards the reaction gas outlet side. thus constituting a means of controlling the gas permeability such that the permeability to the active substances hydrogen and oxygen is low at the reaction gas inlet side and high at the reaction gas outlet side. Since the concentration of the active substances in the reaction gas flowing into the reaction gas passage reduces towards the outlet side of the passages, the uneven distribution of the partial pressure of the active substances that develops at the three-phase interface is compensated by the inverse distribution characteristic of the permeability control means. Since the active substance partial pressure distribution at the three-phase interface can be made uniform, the problems in the prior art that are caused by the uneven partial pressure distribution. such as the uneven distribution of the electric power generating reaction at the three-phase interface, the resulting decrease in the uniformity of operating voltage or current produced in the fuel cell, the inefficient use

of the electrode area and the resulting reduction in electric power generation performance, can be eliminated and a fuel cell provided which makes efficient use of the entire electrode area, and stably provides high electric power generation performance.

4) Brief description of the drawings

Fig. 1 is a sectional view of the gas permeability [control] means of a fuel cell in accordance with an embodiment of the present invention.
Fig. 2 is a sectional view along line A-A of fig. 1.
Fig. 3 is a sectional view along line B-B of fig. 1.
Fig. 4 is a sectional view in accordance with a further embodiment of the present invention.
Fig. 5 is a characteristic curve of the partial pressure distribution of active substances in accordance with an embodiment of the present invention in comparison with the prior art.
Fig. 6 is a perspective view of the general structure of a single cell.

1...Single cell
2...Matrix
3...Fuel electrode
4...Oxidising agent electrode
5,15,35...Ribbed electrode substrate.
6...Electrode catalyst layer
7...Separator
8F,8A.8...Reaction gas
5A.5B,25...Channel (reaction gas passage)
25B...Sloped bottom surface of channel
d...Channel depth
t...Thickness of electrode substrate of channel bottom part